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Physics of Fracture

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The underlying physical bases of present-day fracture theory are examined. It is proposed that the atomically sharp crack should be taken as the cornerstone for modeling propagation processes at the fundamental level. Transmission electron microscopy evidence is presented in support of this contention. Linear continuum fracture mechanics is shown to have intrinsic limitations in its capacity to describe crack-tip phenomena; a more realistic description is provided by lattice statics, incorporating the picture of a crack as a narrow slit terminated by nonlinear linkage bonds. This description establishes a powerful starting point for understanding and predicting the effects of important crack-tip interaction processes. Two such processes, chemically enhanced slow crack growth and process-zone toughening, are discussed in this light. Finally, the nature of strength-controlling flaws in brittle ceramics is considered, with particular reference to the validity of the widely adopted hypothesis that such flaws may be regarded as true microcracks.

I. Introduction

ALONG with our ever-expanding modern-day technology has come an increasing demand for high-performance materials. Thus it is that ceramics, hitherto rejected because of their brittleness, have begun to emerge as attractive candidates for certain engineering applications. Ceramics have qualities, such as high melting points, chemical durability, and intrinsic hardness, which lend themselves to component survival at the extremes of service operating conditions. With this class of materials the chief problem in design is accordingly the containment of potential fracture processes; what properties need to be optimized to guard against the catastrophic formation and growth of cracks?

It is in response to this last question that we have witnessed the evolution of that branch of engineering science known as "fracture mechanics." The formalism of fracture mechanics stems from the basic hypotheses laid down by Griffith in his pioneering paper of 1920[1]: (i) equilibrium extension of well-developed cracks is governed by a balance between mechanical energy released and fracture surface energy gained; (ii) such cracks start from "flaws" in the stressed material. The strength of any given material is determined by both these factors; high "toughness" (resistance to crack extension) and small flaw size are prime requisites for optimal load-bearing capacity. The difficulty with ceramics is that their toughness is inherently so low that they cannot survive operational stress levels if they contain flaws of characteristic dimension 1 to 100 μm. Indeed, with optical fibers, where the operational conditions are unusually stringent, the appropriate flaw dimension may be as low as a few nanometers. What fracture mechanics does is to provide us with a mathematical formalism, based on the picture of a slitlike crack embedded in a linear elastic continuum, for handling the first of the Griffith hypotheses in a general way. Then, given all necessary information on the geometry of the critical flaw in relation to the applied stress field, one has, in principle, the means for evaluating the mechanical response of a component to failure.

For those who concern themselves primarily with the question of *when* fracture occurs, as engineers do, the methodology of fracture mechanics appears to be totally adequate as a predictive tool. However, if we ask ourselves *why* fracture occurs, things start to go wrong. For the critical processes of crack separation must occur at the very tip, and here the linear elastic continuum solutions show singularities. Thus, in inquiring how stresses remotely applied at the outer boundaries of a specimen transmit to the crack tip, there is a limit as to how far we may go with conventional fracture mechanics. The way engineers circumvent this difficulty is to write down empirical crack "laws" for extension in terms of some parameter which characterizes the *intensity* of the locally concentrated stress field, the *distribution* of stresses within the field being taken as invariant. A similar disregard for geometrical details is adopted in the description of flaws for strength analysis; the most common approach is to regard flaws simply as "microcracks," scaled-down versions of true, well-defined cracks, whose characteristic dimensions may be predetermined by empirical testing procedures. In short, engineering fracture analysts concern themselves with the *mechanics,* as distinct from the *mechanisms,* of crack growth.

Clearly, if we wish to understand fracture processes at a fundamental level our attention must turn to the latter aspect. There are two major problems which immediately become apparent.[2] The first of these concerns the assumption of linear elasticity; for a truly Hookean solid the proportionality between stress and strain has no upper limit, implying an infinite strength. Thus the mechanism of material separation at the crack tip is essentially *nonlinear.* The second problem arises in connection with the continuum approximation; the dimensions of the region in which these critical nonlinear processes operate in ceramics are calculated to be small, <1 nm. Hence, the description of separation processes strictly needs to consider the *discrete* nature of matter. What we are effectively saying here is that crack growth is ultimately governed by the complex forces which hold neighboring atoms together in the solid. It is in this context that the title theme of the present paper has its conception.

In what follows an attempt will be made to sketch some of the more important advances in fundamental fracture theory. Inevitably, the selection of topics and the corresponding interpretations will reflect a personal viewpoint. The presentation will focus around one central assertion, that brittle cracks are atomically sharp and propagate by the sequential rupture of bonds. In the first part the justification for making this assertion, and the evidence which supports it, will be given. This will set the scene for modeling two important fracture phenomena in ceramics, slow crack growth due to chemical interactions with environmental species and toughening due to the operation of energy-dissipative pro-

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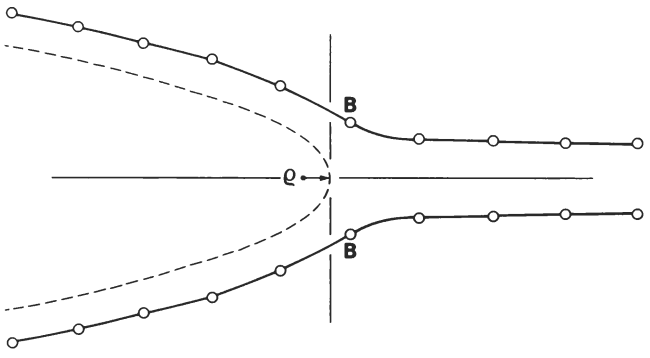

Fig. 1. Tip contours for equilibrium crack in silica glass, as computed from linear elastic fracture mechanics. Crack-tip radius is seen to be significantly smaller than intermolecular spacing.

cesses within the near field of the crack. It will be argued that, by considering events at the sharp tip in isolation from the remainder of the crack system, the potential exists, for the first time, for making *a priori* predictions of propagation laws for particular material systems. Finally, the extent to which small flaws may be regarded as true microcracks will be examined in the light of recent controlled-indentation studies.

II. The Atomically Sharp Crack as the Cornerstone of Brittle Fracture Theory

(1) Intrinsic Limitations of Linear Elastic Continuum Models

We begin with an outline of the now familiar picture of a slitlike crack in a linear elastic, isotropic continuum.[2,3] In accordance with appropriate boundary conditions (i.e. remotely applied tensile loads, traction-free crack walls), the solutions for the near-field stresses and displacements about a crack tip in a material with Young's modulus E and Poisson's ratio ν are of the general form

$$\sigma_{ij}=[K/(2\pi r)^{1/2}]f_{ij}(\theta) \tag{1a}$$

$$u_i=(K/E)\,(r/2\pi)^{1/2}g_i(\theta,\nu) \tag{1b}$$

where r,θ are polar coordinates. The distribution characteristics of the field are determined by the separable coordinate-dependent terms: of these, the radial component is particularly noteworthy, for it shows immediately how the stresses and strains become infinite at $r\rightarrow 0$; the angular quantities $f_{ij}(\theta)$ and $g_i(\theta,\nu)$ are explicit functions which are obtainable from any standard reference source on fracture mechanics (e.g. Refs. 2 and 3). The remaining quantity K uniquely determines the intensity of the field, and is appropriately termed the stress intensity factor. This factor embodies the essential boundary conditions of the crack system; it scales directly with the applied load and is a function of characteristic crack dimensions.

The stress intensity factor is a particularly appealing parameter in engineering mechanics, for, not only does it quantify the driving force on the crack, it satisfies the laws of linear superposition. Consequently, there has developed a strong tendency to formulate crack extension laws exclusively in terms of K. Such laws are of two main types. (i) *Equilibrium* laws, which specify that cracks may extend (stably or unstably) at some critical stress intensity factor, $K=K_c$, which defines the material toughness. In the event that the toughness is determined entirely by the reversible work of free surface creation, γ, it can be demonstrated[2] that $K_c=(2\gamma E)^{1/2}$. Then, coupled with the standard solution for cracks of characteristic length c subjected to uniform applied tensile stress σ, $K=Y\sigma c^{1/2}$ (where Y is a crack geometry term), we obtain an instability condition $\sigma_f=(2\gamma E/Yc_f^{1/2})$, which is the famous Griffith strength formula.[1] (ii) *Kinetic* laws, where, at some subcritical configuration $K<K_c$, the crack can extend at some specifiable velocity, $v=v(K)$. The most important example of kinetic crack growth is that due to chemical interactions with environmental species.

Let us now take a closer look at the crack field solutions at the tip itself. As an illustrative example, consider silica glass for which $E=70$ GPa, $\nu=0.2$, and $K_c=0.75$ MPa·m$^{1/2}$ (as determined from direct observations of crack growth in large-scale test pieces).[4] Figure 1 shows appropriate equilibrium crack-tip contours evaluated from Eq. 1(*b*). The solid contours represent displacements for planes initially separated by one Si–O–Si bond linkage distance across the crack plane, 0.32 nm; the circles along these contours correspond to this same linkage unit, and are to be taken as an indication of the average molecular density rather than of the true atomic structure. It is apparent that the strains ahead of the crack-tip origin are well beyond the Hookean range; the "bond" BB, for instance, has undergone a normal strain of 60%. The dashed contour in Fig. 1 represents the displacements for the initially contacting crack walls. From Eq. 1(*b*) it can be shown that this contour is parabolic with tip radius $\rho=(4/\pi)\,(K/E)^2$; for the equilibrium configuration shown, $\rho_c=0.14$ nm, which is less than one-half the intermolecular separation. Of course, as the crack enters the subcritical region, $K<K_c$, the radius becomes smaller still. It is clear, therefore, that the parabolic crack-tip contour is a physically meaningless concept in terms of the molecular structure; the continuum model cannot be used to describe curvature at the subatomic level. The conclusions drawn here, which can be demonstrated to apply to ceramics in general (at least at room temperatures), suggest that brittle cracks may be more realistically represented as narrow slits terminated at their ends by nonlinear connecting springs of atomic dimensions.

Before pursuing this point in detail it should be pointed out that there exist alternative viewpoints concerning the fundamental nature of crack processes in brittle materials. One of these takes note of the clear evidence for localized plastic zones at crack tips in metals and polymers, and argues that similar zones must exist in ceramics, even if on a submicroscopic scale.[5] Such plastic zones, it is claimed, are necessary to account for the fact that some ceramics have measured toughness values in excess of those expected from surface-energy considerations alone. In this view, fracture is effectively controlled by bulk deformation properties. Another modeling procedure, used extensively by those who study chemically assisted failure, involves the assumption that the crack tip is indeed rounded, and that fracture ensues as a result of some stress-enhanced "sharpening" mechanism (e.g. by preferential dissolution of the fissure walls).[6] Now it is surface chemistry which is the important factor. We should acknowledge here that neither of these two alternatives is totally inconsistent with the sharp-crack concept: for the first, *limited* plasticity (or any other energy-dissipating process) can occur within the near field without altering the essential nature of the sequential bond rupture mechanism; for the second, sharpening may well be feasible, but not beyond the limit of atomic dimensions as already discussed, in which case the model envisaged relates more properly to crack *initiation* from a starting *notch*. It is nevertheless our contention that a truly propagating brittle crack has certain properties that only a nonlinear, atomistic theory can predict, and in this sense the distinctions made above between the different approaches extend beyond the realm of mere semantics.

(2) Direct Observations of Crack Tips and Interfaces: Transmission Electron Microscopy

Until recently, virtually all the evidence cited in favor of one crack-tip model or another in brittle ceramics could be regarded as "circumstantial." Because of the extremely small scale on which the essential separation processes are expected to operate, direct, confirmatory observations have generally lain beyond the reach of ordinary microscopic techniques. (For a survey, see Ref. 7.) However, transmission electron microscopy (TEM), with its ultimate potential for resolving detail at the atomic level (albeit only in solids with a regular, crystalline array), has changed all that. The beautiful work of B. J. Hockey stands alone in this area,[8–10] and here we shall examine some of the findings from his observations which bear on the crack-tip question.

Hockey's studies have been carried out on four select materials, silicon, germanium, silicon carbide, and aluminum oxide, covering a broad spectrum of covalent-ionic bonding. Specimens are indented with a Vickers pyramid to produce the requisite cracks, and are then thinned into a foil, as indicated in Fig. 2(*A*). A typical TEM micrograph of the overall damage pattern produced is shown

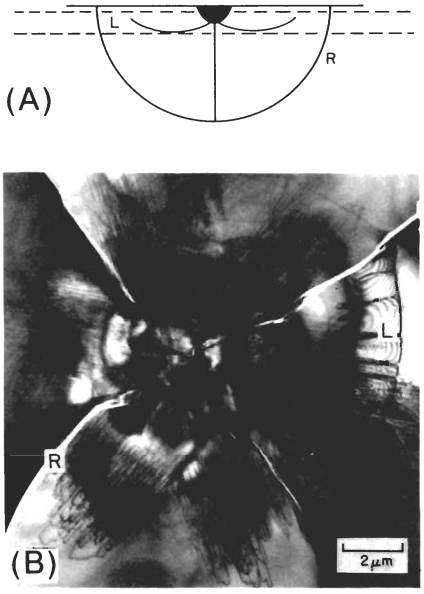

Fig. 2. Transmission electron microscopy of Vickers indentation site. (*A*) Schematic (profile view), showing sections of central deformation zone and associated radial (R) and lateral (L) cracks sampled by foil. (*B*) Corresponding micrograph (normal view) of indentation in SiC. Note intense residual strain associated with deformation center, source of crack-mouth opening displacement (after Ref. 9).

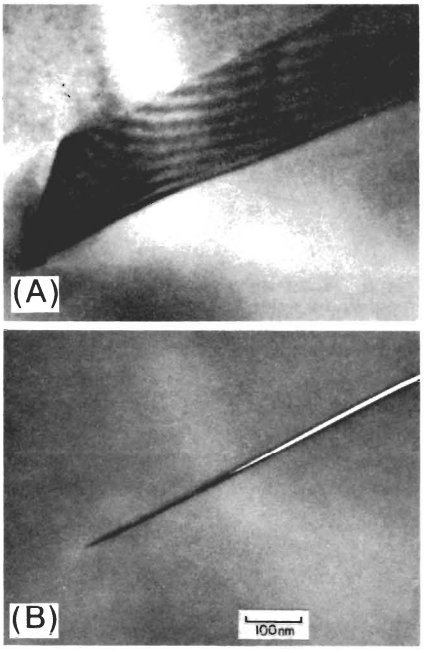

Fig. 3. Transmission electron micrographs of radial crack segment in Si, viewed (*A*) slightly inclined and (*B*) edge-on to crack plane. Note that all diffraction contrast is confined to crack interface (after Ref. 7).

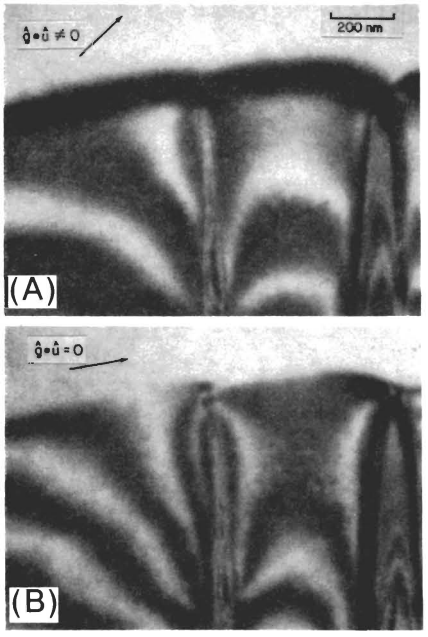

Fig. 4. Transmission electron micrographs of lateral crack segment in Si. Note dark band at crack front, shown (*A*) in contrast and (*B*) out of contrast; $\hat{\mathbf{g}}\cdot\hat{\mathbf{u}}$ visibility criterion, where $\hat{\mathbf{g}}$ is diffraction vector (arrowed) and $\hat{\mathbf{u}}$ is unit vector crack-front normal (not shown), indicates a residual tip-displacement field entirely elastic in character. Distortion of fringe pattern is due to presence of cleavage steps at imperfectly closed interface (after Ref. 7).

in Fig. 2(*B*). The main features of the pattern are the contact deformation zone, characterized by the intense central region of residual-strain contrast, and the two types of crack which emanate from this zone: (i) "radial" cracks, normal to the original specimen surface and thus contained as ribbonlike segments in the foil; (ii) "lateral" cracks, approximately parallel to the specimen surface and contained as relatively extended, near-planar segments.

Let us now look at the tip regions of some of these cracks at higher magnification, Figs. 3 and 4. Figure 3 illustrates a crack segment as viewed in two foil tilt orientations. It is immediately clear that the source of the diffraction contrast is the crack interface itself, about which we shall say more below. The actual crack front is marked by a homogeneous band of contrast, seen to better advantage in the segment in Fig. 4; the systematic disappearance of this contrast under certain reflection conditions indicates a residual elastic strain field about the tip, consistent with the open crack interface seen in the edge-on view of Fig. 3(*B*). Investigation of several hundred indentation sites has shown that the intensity of the contrast band varies considerably from crack to crack; the cracks are wedged open at their mouth by plastic displacements within the central deformation zone, and the portion of this zone sampled by the foil can depend sensitively on the thinning process.[9] Although the evidence for plastic displacements at the crack *mouth* is beyond dispute, not a single sighting has been made at any of the indentation sites examined to suggest that similar displacements occur at the crack *tip*. In terms of the plastic-zone models of crack growth, dislocations (or other elements of plasticity) would be expected to be clearly visible as out-of-plane line defects in micrographs such as those in Figs. 3 and 4.[7] We must therefore conclude that the plastic-zone concept is inappropriate to ceramics at ordinary temperatures.

Let us be clear about our conclusion here. The observations just described do *not* preclude the operation of near-field dislocation sources in all ceramics under all conditions.[7] Indeed, in some softer ceramics at room temperature, and certainly in materials generally at high temperatures, such sources have been clearly identified. What we are asserting is that plasticity cannot constitute the basic material separation process of brittle fracture.

Turning to the interface region behind the tip we find further clues as to the basic crack-tip geometry. In favorable cases, particularly those relatively free of the crack-tip contrast band, interfaces are able to close up to some limited extent and thence heal. The distinction between cracks which have or have not healed is readily made from the diffraction contrast effects[7,9,10]: unhealed cracks are characterized by broad fringes, moire fringes (modulated by thickness extinction contours at surface-inclined interfaces), of the type seen in Figs. 3 and 4; healed cracks are characterized by in-plane, dislocation networks which degenerate from the fringe patterns, as shown in the crack segment in Fig. 5. In both cases the contrast arises from minute degrees of lattice mismatch between diffracting crystal sections on opposite sides of the interface; for instance, a relative lattice rotation of $\approx 10^{-3}$ rad can account for the fringe or dislocation spacing in Fig. 5.[9] The healing

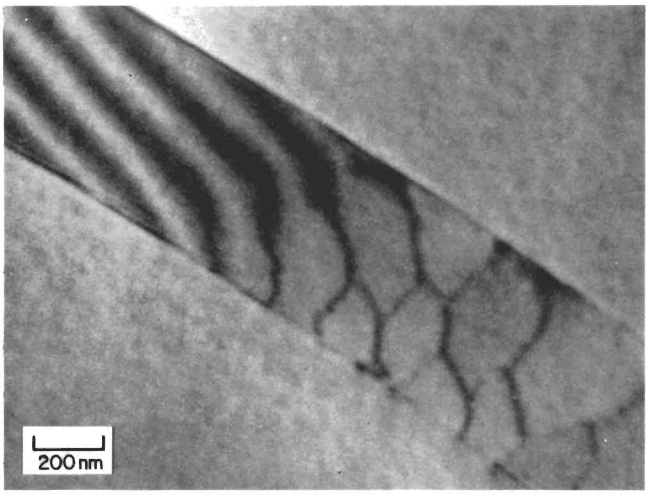

Fig. 5. Transmission electron micrograph of radial crack segment in Al_2O_3, showing interface behind tip. Note how moire fringes degenerate continuously into network dislocations (after Ref. 7).

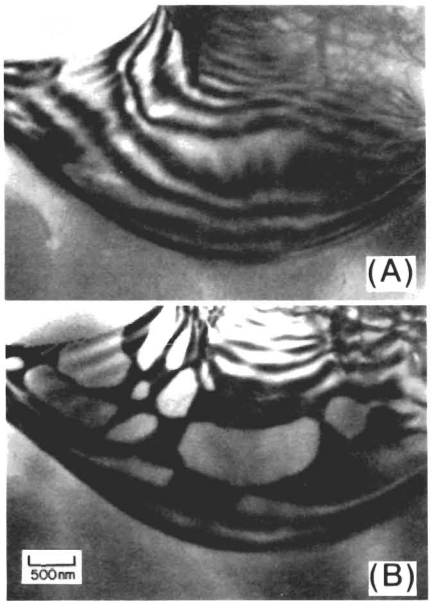

Fig. 6. Transmission micrographs of lateral crack segment in Al_2O_3, showing same interfacial portion under invariant diffraction conditions (*A*) on initial exposure and (*B*) after prolonged exposure to electron beam. The change in pattern can be explained in terms of a stacking fault decomposition process (after Ref. 10).

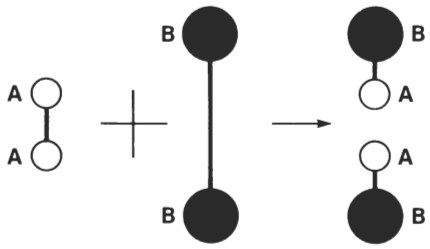

Fig. 8. Schematic of chemically induced bond rupture, considered in isolation from matrix lattice. As portrayed, the reaction can be considered as one between diatomic molecules.

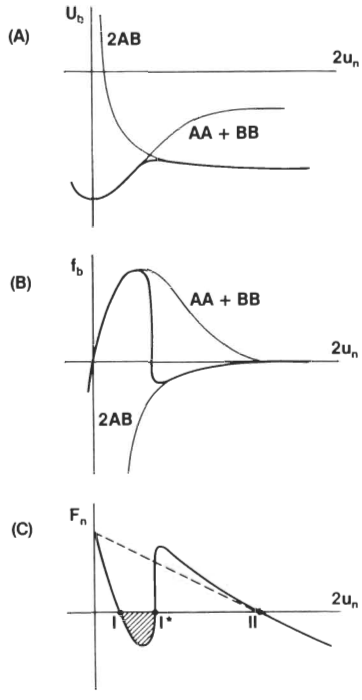

Fig. 9. Interatomic interaction diagrams for crack-tip bonds subjected to chemical process depicted in Fig. 8. (*A*) Potential energy as function of reaction coordinate for bond B–B separation. Heavy curve denoting minimum energy configuration indicates sudden transition from unreacted to reacted state at critical separation. (*B*) Corresponding restoring force diagram. (*C*) Composite crack-tip bond-force plot, combining (inverse) bond restoring curve with linear bias of applied load plus lattice constraint (inclined dashed line) (after Ref. 18).

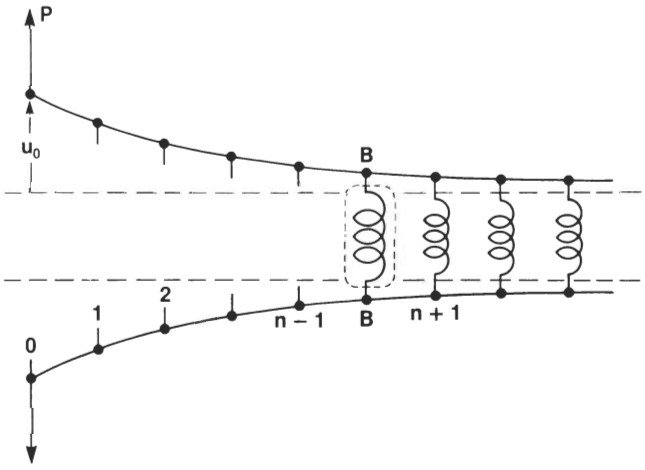

Fig. 7. Quasi-one-dimensional model of a sharp crack, with nonlinear crack-tip bond BB embedded in a linear "lattice" (after Ref. 13).

configuration is simply a relaxed version of its unhealed counterpart, in which atoms have recontacted and bonds realigned to the maximum extent possible, commensurate with the mismatch. The restoration may be somewhat incomplete, however, as can be seen by the weird and wonderful configurational changes that may be induced in certain instances by electron-beam heating. Figure 6 is one such instance.[10] Now the fact that healing of this type, either spontaneous or thermally induced, occurs in brittle solids at all, and that such healing is most commonly observed in the region immediately adjacent the crack front, appears to rule out any crack-tip concept based on an irreversible surround zone or on an intrinsically rounded tip. In neither of these last two cases would a freshly propagated crack be able to close up at any point along its interface, let alone heal.

The electron microscopy evidence concerning reversibility in the basic mechanism of brittle fracture accordingly imposes certain requirements in crack modeling. The picture of an atomically sharp slit meets all these requirements. In this context the image conjured up by Barenblatt[11] of the brittle crack as a "zipper," freely running and closing (i.e. without dislocation emission), is appropriate.

It could be argued that the above electron microscopy observations relate to only one special kind of crack, that produced in controlled contact experiments. However, it has been extensively demonstrated that contact cracks follow the same growth laws as other crack configurations used in more conventional mechanical testing arrangements[12] (see e.g. Section IV (2)). The conclusions would therefore seem to be quite general.

(3) Discrete Lattice Models

We have argued strongly for modeling brittle cracks as slits terminated by linkage bonds. According to this notion it is hardly appropriate to seek fundamental descriptions of fracture processes in terms of properties of the bulk, or even of the surface; our crack is effectively a line defect. Then again, as with the analogous case of the dislocation, it is not necessary for this line to move as a unit; rather, bonds can be broken individually along the front, causing atomic-scale jumps ("kink" motion). We shall be looking at an approach which allows one to decouple the equations governing the critical crack-tip bonds from the remainder of the system, thus reducing the problem to one of interatomic force laws.

The mathematical vehicle for achieving this end is lattice statics. Lattice modeling has been developed to a high level of sophistication by Thomson and coworkers.[13–15] Several variant lattice representations of cracks have been considered by these and other workers. We shall focus our attention on the simplest; the chief objective here is to draw general conclusions with minimum geometrical or mathematical complication. This approach sacrifices structural reality for physical insight; it allows for analytical solutions of an otherwise intractable lattice problem. The alternative is to set up a

"realistic" interatomic potential for a three-dimensional crystal and simulate the crack system on a computer. While such exercises can be useful, for instance in confirming the atomic sharpness of brittle cracks,[16] the conclusions drawn are generally limited to the specific system under consideration.

Consider, then, the atomistic crack model shown in Fig. 7.[13] The "atoms" are point masses and the "bonds" which link them are springs. The spring elements are of two types, stretchable (transverse) and bendable (longitudinal). These element types are assumed to be linear in their force/displacement response, with elastic constants α and β, respectively, except for the nth stretchable bond at the crack tip, which is *necessarily* nonlinear. Behind the tip the bonds are of course "broken," i.e. they are stretched beyond their range of interaction. The entire configuration is maintained in equilibrium by the application of opening forces P at the crack mouth. Despite its unlikely appearance, this model contains all the essential features of a brittle crack: it has the quality of an atomically sharp slit; it has nonlinearity built in at the very point where separation is to occur; and the system as a whole is nevertheless linear in its elastic behavior, with provision for incorporating rigidity as well as stiffness characteristics.

The procedure for determining the equilibrium-displacement solutions for the configuration in Fig. 7 is straightforward, if tedious. One begins with an expression for the potential energy U of the entire system, expressed in terms of the displacements u_j for all atom pairs, i.e. $j=0,1\ldots\infty$. For all atom pairs other than that at the crack tip the energy terms are harmonic; for the nth atom pair the energy term involves the unknown nonlinear force function $f_b(u_n)$ (the area under the force/displacement curve providing the requisite energy function). Now for equilibrium at each atom pair the condition $\partial U/\partial(2u_j)=0$ must be met (note $2u_j$ is the total bond displacement). Analytical solutions of the functional form $u_j(P,n,u_n)$ are then obtainable for all but the crack-tip bond, i.e. for all $j\neq n$. These solutions may be combined linearly in such a way as to reduce the system potential energy to an expression for U_n in terms of crack-tip displacement. The requirement that the crack-tip bond must itself be in equilibrium gives the final result

$$\begin{aligned} F_n &= -\partial U_n/\partial(2u_n) \\ &= P(1+n/\zeta)-(\zeta-1)\alpha u_n - f_b(u_n) \end{aligned} \tag{2}$$

where $\zeta=\zeta(\alpha/\beta)$. Equation (2) conveniently defines a generalized force for crack-tip bond rupture; for $F_n>0$ the bond opens, for $F_n<0$ it closes.

This result has important implications in the modeling of the micromechanics of fracture. Each of the separable terms in Eq. (2) represents a distinctive source of driving or retarding force on the crack. The first term represents the applied driving force, the second represents the retarding force due to the constraint of the linear elastic lattice surrounding the crack tip, and the third represents the restoring force due to the stretched crack-tip bond itself. It is apparent, therefore, that the problem of specifying a suitable interatomic force function $f_b(u_n)$ for a given material system can be handled quite independently of the remainder of the crack system. That is to say, the intrinsic response of the crack-tip atom pair may be considered in isolation, and folded into the crack-force equation after an appropriate solution is found. In this view the ultimate answers to the brittle fracture question are to be sought in the properties of the chemical bond.

We should note that the lattice statics viewpoint does not conflict with the thermodynamic arguments of Griffith. Although our cracks have the essential character of line defects, the end result of the motion of such defects is indeed the creation of new surface. Thus for equilibrium configurations we should expect the lattice solutions in the continuum limit of bond distance $\to 0$ and $n\to\infty$ to yield crack resistance terms in the intrinsic surface energy.[15]

III. Applications of the Sharp-Crack Concept

(1) Crack-Tip Chemistry

One of the most important fracture phenomena in the strength of ceramics is that of slow crack growth. As mentioned in Section II, this is a kinetic process, enhanced dramatically by suitable chemical environments. For a given material/environment system it is possible to determine an empirical crack velocity function $v(K)$, this function generally increasing monotonically, and steeply, as K increases. The reason for the importance of slow crack growth lies in the fact that it can cause initially small cracks to grow, in time, to catastrophic sizes. Specification of an appropriate crack velocity function is accordingly a key factor in modern-day mechanical designing with ceramics.

Yet our fundamental understanding of chemical interactions with cracks has been almost totally lacking. It is true that some progress toward a mechanistic description has been made using the theory of reaction rates in conjunction with continuum mechanics descriptions of tip geometry,[17] but this type of approach is somewhat phenomenological; it can explain how certain variables, such as temperature and concentration, enter the problem, but is limited in its capacity to use data from one system to predict the response of another. Indeed, our current knowledge of which environments are most likely to interact with which materials is totally empirical. Why is it, for instance, that water is such a potent crack-growth agent in some, but not all, glasses and ceramics?

It is the contention here that the answers to questions like this should be sought at the molecular level. We reemphasize that in Eq. (2) the nonlinear bond-force term $f_b(u_n)$ is a separable component of the overall crack-driving force. The question then arises as to what form this function must take in the event of a chemical interaction at the crack tip. To illustrate, we investigate the relatively simple interaction shown in Fig. 8, in which an environmental molecule A–A combines with the crack-tip bond –B–B– to produce terminal bonds A–B–.[18] Insofar as such events may be treated in isolation from the remainder of the crack system, the rupture process may be considered in terms of the well-studied reaction AA+BB→2AB between diatomic gas molecules.[19] This reaction can be represented schematically on interatomic potential and force diagrams, as in Fig. 9:

(i) In Fig. 9(*A*) the potential energy U_b is plotted as a function of bond B–B separation for the unreacted and reacted states, the molecule AA being allowed at all times to occupy a position of equilibrium. This diagram tells us that as the crack-tip bond is progressively stretched there is a critical separation beyond which the reacted state becomes energetically more favorable, i.e. the bond is chemically "broken." The depth of the potential barrier to bond rupture is therefore lowered considerably by the presence of the environmental species.

(ii) Figure 9(*B*) is an equivalent, force representation of this same interaction, generated from the definition $f_b=\partial U_b/\partial(2u_n)$. In this plot the rupture point is dramatically evident as the critical separation where the bond force switches from attractive to repulsive.

(iii) Finally, Fig. 9(*C*) is a composite plot of all three force terms in Eq. (2), showing how the applied loading and lattice constraint bias the (negative) contribution of the bond to the total crack opening force. Such diagrams are useful for determining the relative importance of the force components as the bond is taken from stable configuration I (bond intact) to stable configuration II (bond broken) via the activated complex state I*.[18] Suffice it to say that the model introduces in a perfectly natural way the one essential ingredient of any rate process, that of an energy barrier, as the shaded area bounded by states I and I* under the force/separation curve. (Note that in Fig. 9(*C*) this forward-motion barrier is shown smaller than its backward-motion counterpart, determined as the area bounded by states II and I*, corresponding to healing by desorption.) With this barrier determined, crack velocity functions can in principle be computed from the theory of thermal fluctuations using the usual methods of statistical mechanics.[20]

The scheme presented in Fig. 9 is, of course, oversimplistic. Quite apart from the fact that the crack-tip bond is embedded in a totally unrealistic structure (Fig. 7), it is implied that the intrinsic energy curves for the unreacted and reacted crack-bond states (Fig. 9(*A*)) are readily accessible for specific systems. Unfortunately, this is not so; this is the province of quantum chemistry, where the energy states of the most elementary reactions are barely understood. Nevertheless, the approach serves well as a basis for making some qualitative predictions, as we shall demonstrate using

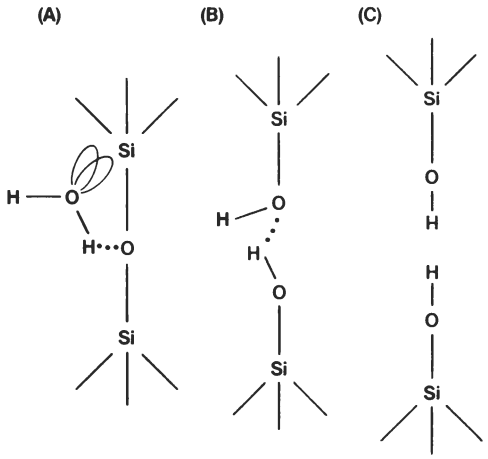

Fig. 10. Interaction between water molecule and strained crack-tip bond in glass. Stages represent (*A*) adsorption, (*B*) reaction, and (*C*) separation. Note that the process involves no material removal from surface (after Ref. 21).

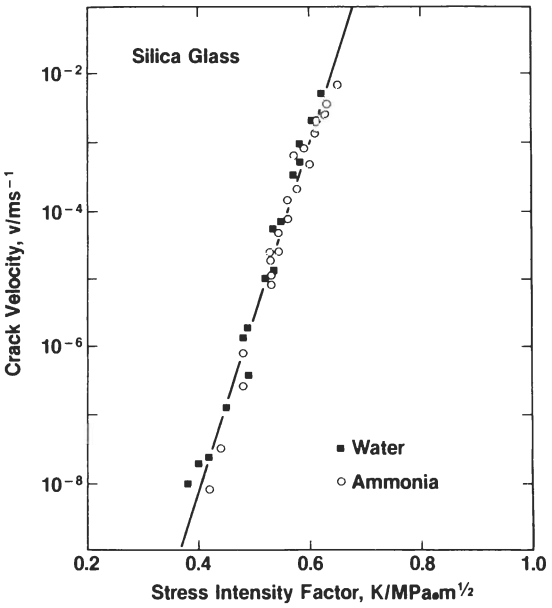

Fig. 11. Crack velocity measured as function of stress intensity factor for silica glass in water and ammonia. The line through the data is simply an empirical fit (after Ref. 21).

some impressive results from a study of fracture kinetics in silica glass by Michalske and Freiman.[21]

What Michalske and Freiman[21] have done is to address the molecular interaction problem from an electron orbital viewpoint. They begin by inquiring why it is that silica glass is so susceptible to slow crack growth in the presence of water. It is envisaged that the incoming water molecule interacts with the Si–O–Si crack-tip bond in three stages, Fig. 10:

(i) Step (*A*) involves attachment and alignment of the water molecule with the bridging bond. An important facet of this step is the electronic structure of both interacting members. The water molecule may be thought of as having an approximately tetrahedral electron orbital configuration (largely sp^3 hybrids) about the central oxygen atom; two of these orbitals form bonds with the hydrogen atoms and the remaining two form lone-electron pairs. This results in a polar disposition in which there is a net negative charge at the lone-pair end of the molecule and a corresponding positive charge at the hydrogen end. The crack-tip bond also has some polar character, with net positive charge on the silicon atoms. Hence the "adsorption" configuration shown.

(ii) Step (*B*) defines the reaction stage, in which the Si–O–Si bond has been stretched to the critical crossover point on the energy curve of Fig. 9(*A*). At this point the water molecule donates an electron to the silicon, and a proton to the oxygen, in the stretched linkage unit. This transfer produces two new bonds, one between the original silicon and the oxygen from the water, the other between the original oxygen and a hydrogen.

(iii) Step (*C*) involves severance of a weak hydrogen bond in what remains of the initial water molecule after electron redistribution. Bond rupture is now complete, and the fracture surface is saturated with hydroxyl groups.

The importance of the Michalske-Freiman[21] approach lies in its facility to make predictions, for the first time, as to the susceptibility of alternative material/environment systems to kinetic fracture. According to the above description, the essential ingredients for strong interactions are the capacity for the incoming molecule to donate both electrons and protons and a degree of polarity in the crack-tip linkage bond. To confirm the necessity of the first of these ingredients, Michalske and Freiman conducted comparative $v(K)$ tests on their silica glass, using ammonia as a second environmental species. They pointed out that the ammonia molecule has a similar electron orbital structure to that of water, except that now the central nitrogen forms bonds with three hydrogens, leaving a single lone-pair orbital. Their experimental data, illustrated in Fig. 11, indicate that both test molecular species are effective in promoting slow crack growth. On the other hand, molecules without *both* lone-pair orbitals and hydrogens (e. g. carbon monoxide) proved to be completely ineffectual. As to the second ingredient, the authors cited the fact that covalent structures, such as monocrystalline silicon, are notoriously immune from rate effects in fracture.

(2) Toughening: Process Zone and Enclave Models

We have repeatedly alluded to the brittleness of ceramics. One of the objectives of materials scientists working with ceramics is accordingly to find ways of improving the toughness, K_c. Basically, this boils down to designing the material microstructure so that the external work of propagating cracks can be dissipated in other than mere surface creation. Thus arises the idea of a "process zone," a concentrated, highly stressed volume about the crack tip within which discrete "energy sinks" are activated. These sinks may be in the form of second-phase particles which undergo phase transformations[22] or localized microcracking,[23] or incipient dislocation loops which expand or contract without in any way "blunting" the crack tip.[24] Given that the operation of such processes can lead to substantial toughening, perhaps several times greater than the intrinsic surface energy contribution, how must we modify the crack-tip modeling to accommodate additional factors in the system energy balance?

This last question raises subtle issues which have in the past been improperly addressed in the scientific literature. Among these is the issue of mechanics vs mechanisms mentioned earlier (Section I). It is often stated that, in cases where the surface energy makes a relatively small contribution to the toughness, it may be neglected in any consideration of general crack-growth laws: In effect, this is tantamount to saying that the dissipative elements within the process zone are actually responsible for separating the material at the tip. However, if it is acknowledged that these dissipative elements need not interact with the crack in such a way as to produce physical changes in the tip structure, premises which throw away the bond rupture resistance terms can lead to serious misconceptions in the understanding of fracture behavior. For, if the nature of the crack tip remains invariant the extension must be governed by the intrinsic cohesive properties, as before, and the role of the process zone simply becomes one of "shielding" the crack tip from the remotely applied loads. In this description the cohesive term can exert a profound influence on the overall fracture criterion, by controlling the scale of the process zone.

It is as a result of this type of thinking that "elastic enclave" models have recently been developed.[25–29] The basic scheme is shown in Fig. 12. Within the enclave the deformation is entirely elastic (with nonlinear components, of course) and the crack extends in accordance with the fundamental laws of interatomic

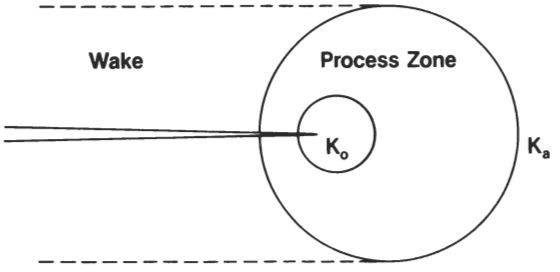

Fig. 12. Enclave model of sharp crack. Region between two circles about crack tip denotes process zone, within which energy sinks operate; as crack advances, process zone leaves a "wake" of "transformed" material. Elsewhere the field is elastic: Outer circle denotes limiting radial distance at which stresses are sufficient to activate the sinks; inner circle denotes effective homogeneous enclave, determined approximately by mean spacing between sinks. Process zone "shields" enclave from remotely applied loads, so that inner and outer elastic regions experience different K fields.

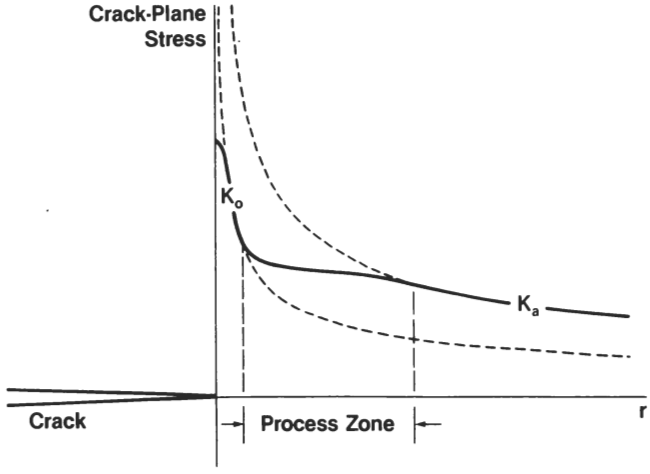

Fig. 13. Distribution of normal crack-plane stresses ahead of tip, showing shielding effect due to relaxation processes within process zone. Outer and inner elastic regions are characterized by well-defined K fields (notwithstanding that in the latter case a stress cutoff must occur due to finite cohesive strength); whereas K_a is quantity measured in fracture test, K_0 governs crack extension.

bonding, exactly as if it were in a perfectly brittle solid. However, the K field appropriate to the enclave can be quite different to that which would apply in the brittle ideal. The effect of introducing the surrounding process zone is to screen the enclave from the outer, linear elastic region, so that the crack-tip K field is reduced relative to the value determined from the level of the applied loading, Fig. 13. The greater the screening, the greater must be the applied loading to maintain a specified crack-extension condition at the tip.

In this description a complete analysis of crack extension requires two sets of equations: the first of these, expressible in terms of the crack-tip field intensity, K_0, has its roots in the intrinsic bonding properties, as already discussed; the second relates this field intensity to the corresponding quantity K_a for the outer field in terms of appropriate process zone parameters. Following the course laid down in the previous sections of this paper we may write the first set in the form

$$K_0^* = (2\gamma E)^{1/2} \tag{3a}$$

$$v = v(K_0) \qquad (K_0 < K_0^*) \tag{3b}$$

where the asterisk denotes a critical equilibrium configuration. Herein is contained the mechanistic component of the overall fracture condition. The second set of equations derives from purely mechanical considerations and as such will not receive detailed attention here. They may be expressed most simply as

$$K_a^* = K_c \tag{4a}$$

$$K_a = K_0 + K_p \tag{4b}$$

where K_c is the contribution of the process-zone screening to the apparent stress intensity factor. The key to the problem then rests with a computation of K_c in terms of the energetics and density distributions of the dissipative sinks. Since this quantity will generally depend in turn on the field intensity within the enclave, i. e. $K_p = K_p(K_0)$, it is clear that Eqs. (3) and (4) can be strongly interdependent.

This interdependence can be illustrated in a qualitative way, without reference to a specific energy dissipative process, in the manner of Fig. 14. Simplistically, the plots in this figure may be seen as the manifestation of a K shift, in accordance with Eq. (4*b*). Thus the vertical dashed lines, representing the critical stress intensity factors for the inner and outer elastic regions, indicate the "degree of toughening" $K_a^*/K_0^* = K_c/(2\gamma E)^{1/2}$ associated with the introduction of the process zone. The solid inclined lines represent the corresponding crack velocity functions $v(K_0)$ and $v(K_a)$, the latter being generated by arbitrarily choosing $K_c \propto K_0$. (In view of this arbitrariness, and of the fact that the axes in Fig. 14 have been plotted logarithmically to emphasize the general steepness of the velocity functions, no special significance should be attached to the appearance of the lines as parallel, or even straight.) The importance of this diagram is that it shows the great sensitivity of the velocity response to process zone mechanics; small amounts of toughening, say less than a factor of two, may lead to velocity reductions of several orders of magnitude. Such effects could be highly beneficial in the design of ceramics, especially where long lifetimes constitute a prime requirement.

By contrast, crack-tip models which assume that the processes of toughening account for the mechanisms of separation as well as the mechanics of stress intensification would predict no such sensitivity of velocity response, since the environmental species responsible for the kinetics do not have direct access to the sinks. Definitive evidence on the matter appears to be lacking in ceramics, although the mounting conviction that cracks may remain atomically sharp even in some metals with comparatively gross crack-tip zones[25,30] lends strong credance to the shielding concepts expressed in Fig. 14. Systematic crack velocity studies on ceramic test specimens which have been subjected to varying degrees of toughening could be most revealing.

IV. Micromechanics of Flaws

(1) Crack Evolution From Flaws: Propagation or Initiation?

Most fundamental fracture studies are conducted on specimens containing well-defined cracks which can be followed at all stages of their growth. In practical strength tests on brittle ceramics, however, no such element of control is generally possible. The flaws responsible for failure are in most instances too small for direct in situ observation, and in any case the location of the critical site in a large flaw population cannot usually be predetermined. In the absence of definitive evidence on the nature of flaws, the fracture mechanics approach has been to treat them as "equivalent microcracks," miniature entities which obey the same growth laws as their supposed macroscopic counterparts. Indeed, this approach has become central to the design philosophy for ceramics. Nevertheless, there is an alternative viewpoint which argues that flaws do not have the essential character of sharp microcracks; the focus in the failure micromechanics then shifts from the propagation to the initiation of cracks. This viewpoint can lead to different predictions in strength characteristics and, as such, deserves consideration.

Somewhat ironically, the main contenders as controlling mechanisms for crack initiation from flaws are precisely those we have rejected as pertinent to crack propagation, concentrated plasticity, and notch sharpening. This time there is some supportive evidence for both these mechanisms in ceramics, although exactly how widespread each is in the characterization of strength properties remains to be established. The basic requirement of all such models of this type is that there should exist a means by which a local stress concentration can be built up to the point where the cohesive

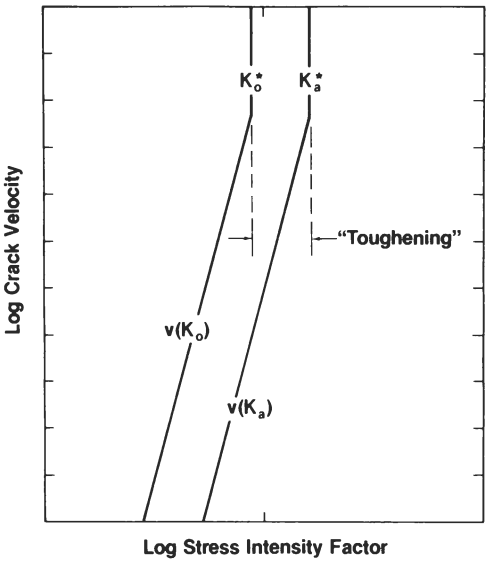

Fig. 14. Schematic representation of crack velocity functions, plotted in terms of both outer and inner K field parameters. Axes plotted logarithmically, in arbitrary units, with decade markers to indicate typical ranges of variation. Note that small toughening effect due to operation of process zone, corresponding to shift from K_0^* to K_a^*, can cause relatively large suppression of kinetic crack growth.

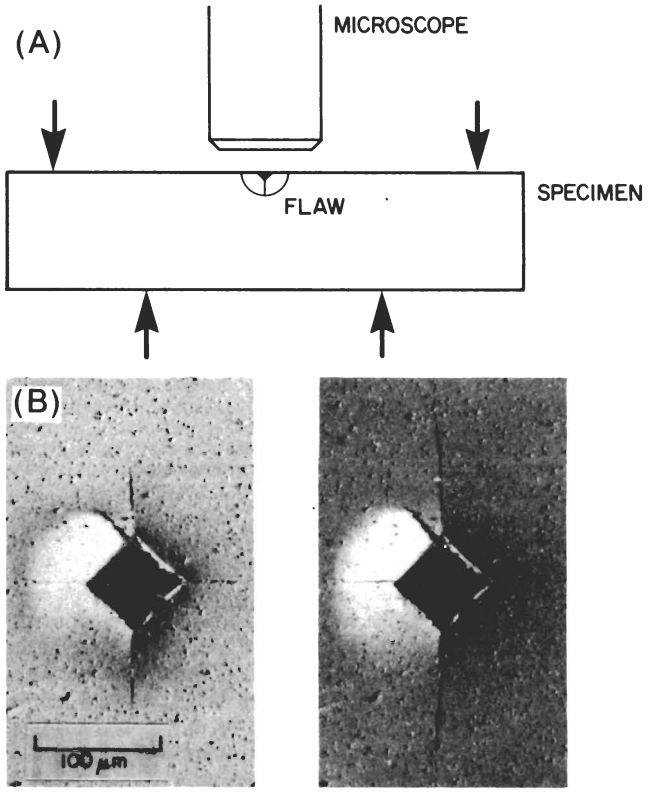

Fig. 15. Growth of radial cracks at indentation flaws during strength testing. (*A*) Schematic of experimental arrangement for viewing cracks in situ in 4-point bend test. (*B*) Micrographs showing crack extension in Vickers-indented hot-pressed Si_3N_4 as bending load is applied. Such observations are clearly inconsistent with any "blunt flaw" hypothesis based on dominance of flaw sharpening over lengthening (courtesy D. B. Marshall).

strength is locally exceeded. Such a buildup ought to be manifest in delayed failure experiments as an *incubation* period prior to any true crack growth. Studies of the strength of silicate glasses after acid-immersion treatments have revealed apparent incubation phenomena,[31,32] although again no direct observations of the initial flaw could be made. Inability to observe flaw response directly has not, however, dissuaded one school of workers from constructing detailed theories of fatigue in silicate glasses from empirically based premises concerning the geometry of "rounded crack tips" and the rate laws which apply at these tips; insofar as they suggest that sharpening completely dominates lengthening in the flaw "growth" history,[33] such theories strictly fall outside the broad compass of the fracture mechanics formalism.

The point that we would make here in the context of the above discussion is that there is an onus on the proponents of any flaw theory to provide definitive evidence in support of their underlying assumptions, or else to identify the limitations of their modeling. Attempts at self-critical analysis have not been altogether common in this area of scientific endeavor, a result of which has been considerable confusion by some workers in their efforts to gain a theoretical understanding of their fracture data. One such example is the adoption of the rounded-contour concept, which we have acknowledged as having some plausibility in the description of certain flaw types, to explain the growth characteristics of well-defined cracks. We would not argue that large-scale fissures with rounded tips are figments of the imagination; indeed, there is indisputable evidence to show that cracks, however sharp in their initial growth, can be readily "blunted" by chemical attack, annealing, etc. However, such entities differ in one important respect from truly brittle cracks, in that their micromechanics are governed by effective contour radii rather than by bonding distances, i.e. they have the quality of notches. The growth laws for "blunt cracks" are no longer uniquely expressible in terms of parameters of the K field,[34] contrary to the general fracture mechanics experience.

(2) *Controlled Indentation Flaws*

One way of investigating some of the issues raised above is to introduce critical flaws in a deliberate and controlled manner, such that direct observations can be made of the evolution to failure. The technique which has received most attention recently is that of sharp-point indentation. A standard Vickers or Knoop diamond pyramid indenter is most commonly used, the load on which determines the scale of the flaw. At relatively high loads well-developed radial cracks (recall Section II (2)) are readily identifiable at the impression corners. On application of a tensile stress these cracks can be seen to undergo significant extension prior to failure, as in Fig. 15.[35] Moreover, provided due account is taken of all driving forces on the system, including residual stress terms associated with the central deformation zone (Fig. 2), the cracks propagate in accord with the established fracture mechanics laws for large-scale test pieces, under both equilibrium[35] and kinetic[36] failure conditions. Since these same indentation cracks have been shown to provide a close simulation of surface damage in the particle impact,[37] cutting,[38] and machining[39] of ceramics, the results to be discussed here may be considered to have a certain generality.

One particular series of Vickers indentation tests on the fatigue properties of silicate glasses in water[40,41] bears strongly on some of the questions previously raised concerning the nature of flaws. In these studies the contact load was used as a variable to adjust the scale of the critical flaw in each prospective strength test piece. This way the scale of the process could be progressively diminished, and the universality of the kinetic crack laws thereby investigated down toward submicroscopic dimensions. The theoretical framework for analyzing the data stems from two basic starting relations, one expressing the net stress intensity factor for the radial crack in terms of residual and applied driving-force components and the other expressing the crack velocity as a power-law function. The resulting fatigue equation for the failure stress σ_f in terms of the lifetime t_f has the familiar form[42]

$$t_f \sigma_f^{n'} = \lambda' \tag{5}$$

except that now the parameters designated by primed notation are controlled by the indentation conditions; for Vickers indentations these parameters have the functional dependence $\lambda' = \lambda'(P) \propto P^{-(n'-2)/3}$, where P is the peak contact load, and $n' = n'(n)$, where n is the true crack velocity exponent. Accord-

ingly, a logarithmic plot of $t_f\sigma_f^{n'}$ vs P should yield a straight line, thus providing a convenient graphical scheme for evaluating the size effect in the fracture mechanics.

The results obtained from the fatigue studies in Refs. 40 and 41 are appropriately plotted in Fig. 16. In this plot the data points fit the linear prediction from macroscopic crack laws over only a portion of the indentation load range; below a threshold load the data points show an abrupt jump (corresponding to an increase in lifetime at any specified service stress). Microscopic examination of the indentation sites prior to the fatigue testing identifies this threshold as that for radial crack "pop-in," defining a level below which the hardness impressions appear to be free of any microcracking.[43] Nevertheless, even in the subthreshold region the data plotted in the figure represent failures from the indentation sites, so it is evident that the transition is one from crack propagation to crack initiation. In terms of theoretical understanding this is a transition from the well-defined realm of fracture mechanics (as embodied in Eq. (5)) to the muddy waters of flaw micromechanics (with all the uncertainties and limitations alluded to in Section IV (1)).

The important conclusion to be drawn from Fig. 16 is that there exists considerable danger in extrapolating macroscopic crack laws into the domain of submicroscopic flaws. Although we have given explicit attention here only to indentation flaws, independent studies on other systems, e. g. those associated with inclusions or second-phase particles,[23] show analogous threshold microcracking behavior. Indeed, size effects of this type may be the rule rather than the exception; the crack formation process has the hallmark of a classical nucleation and growth phenomenon, namely the expenditure of volume energy (elastically stored about the flaw center) in favor of surface energy (associated with the crack interface). There are clear implications here for those who seek to design in the ultrahigh-strength region; the unconditional application of fracture mechanics principles carries an element of danger, particularly in the prediction of parameter-sensitive properties such as lifetime. Optical fibers provide just one example of materials evaluation in which fracture mechanics has been adopted extensively with little or no attempt to identify and understand the underlying sources of failure. Exploration of the subthreshold domain of flaw response must surely constitute a rich area of future research for ceramics scientists.

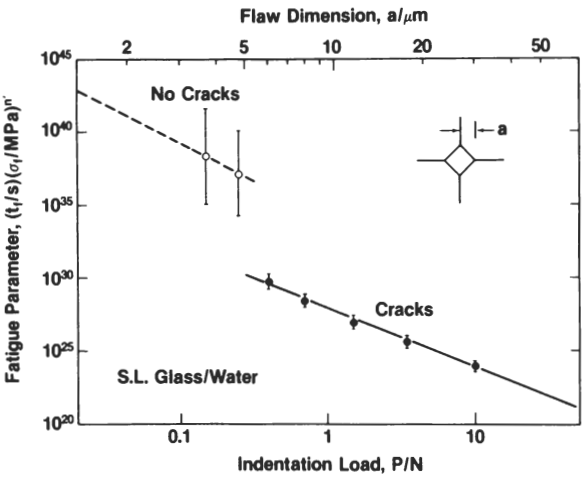

Fig. 16. Fatigue plot for soda-lime glass in water, showing variation of results with indentation load P. (Note that abscissa may be plotted in terms of equivalent flaw dimension a, using hardness $H=P/2a^2=5.5$ GPa for Vickers indentations on glass as the basis for conversion.) Data points from Refs. 40 and 41. Plotted in accordance with macroscopic crack-law parameters, from which values of n' used in ordinate and solid line prediction of Eq. (5) are evaluated. Dashed line through subthreshold data is empirical fit only.

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